Haldane Gap in Three Dimensions: A Rigorous Example

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A quasi-one-dimensional $S=1$ antiferromagnet with XXZ-type intrachain couplings and Ising-type interchain couplings is studied. It has been suggested that, when the interchain couplings are sufficiently small, such a system has no long-range order even at $T=0$ because of strong quantum fluctuations caused by the Haldane gap. Treating the problem in a restricted Hilbert space which describes the low-energy behavior of $S=1$ antiferromagnets rather accurately, we prove two theorems which establish the existence of a unique disordered ground state with a gap.

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Haldane[1] made a fascinating prediction that, in a spin-S Heisenberg antiferromagnetic chain, the ground state is massive and accompanied by a finite energy gap when S is an integer, while the ground state is massless and there is no gap when S is a half odd integer. Haldane made use of a field-theoretic argument based on the large-S limit, so initially it was not clear whether the conclusion applied to small values of S such as $S=1$. By now there have appeared numerical,[2] experimental,[3,4] and rigorous works[5,6] which support the prediction even for $S=1$.

All of the systems used in the experimental works to observe the Haldane gap have been the quasi-one-dimensional (1D) systems which are collections of chains forming three-dimensional (3D) lattices. Since they have strong intrachain couplings and small interchain couplings, they are expected to behave as 1D systems at temperatures not too low. But as the temperature is lowered, it is usually expected that the 3D nature eventually dominates the system and long-range Néel order takes place. This is the case in the good $S=1$ quasi-1D antiferromagnet $CsNiCl_3$ which Néel orders at $T_N \cong 4.9$ K, and in which the indications of the Haldane gap are observed above T_N.[3]

In $Ni(C_2H_8N_2)_2NO_2(ClO_4)$ (abbreviated NENP)[4] which is also a good $S=1$ quasi-1D antiferromagnet, however, no Néel order has been observed down to 1.2 K. Contrary to the general belief in the "universality," it was suggested that the system does not order even at $T=0$. Prior to the experimental result, Kosevich and Chubukov[7] noted that the ground state of a quasi-1D $S=1$ antiferromagnet may be disordered when the interchain couplings are sufficiently small. However, their reasoning is a naive perturbative argument which, in my opinion, is not reliable. Affleck's elegant (but approximate) field-theoretic analysis in the $S\rightarrow\infty$ limit,[8] and Sakai and Takahashi's interchain mean-field theory[9] lead to similar conclusions.

It is well known that a *finite* system with a unique ground state and a gap is generally stable under a small perturbation. One might then imagine that small interchain couplings in a quasi-1D $S=1$ antiferromagnet may be regarded as irrelevant perturbations since the unperturbed 1D system has a gap. This is essentially the argument made by Kosevich and Chubukov.[7] But we stress that the present problem is much more subtle and deeper. First of all one should realize that the perturbations in this case are applied over the infinitely large 1D system. The total perturbation energy is always infinite, and there is no simple reason for the finite Haldane gap to survive the perturbation. Indeed, perturbations with certain long-range coherence (like the staggered magnetic field) alter the long-range behavior of the system quite drastically, no matter how small their magnitude is. Moreover, one has to note that a quasi-1D system can also be regarded as a collection of infinitely many 2D spin systems interacting through strong interlayer couplings. Since each 2D layer itself has the ability to exhibit long-range order, there is no guarantee that they do not cause some nonlocal or nonperturbative effects.[10] To settle the problem, we need an essentially nonperturbative analysis which can deal with the competition between the quantum fluctuations caused by the Haldane effect and Néel order favored by the 2D subsystems.

From a theoretical point of view, the possibility of 3D antiferromagnets with disordered ground states has already been established rigorously. Affleck, Kennedy, Lieb, and the present author[6,11] constructed a class of valence-bond-solid Hamiltonians in three and more dimensions which have disordered ground states. The similar possibility in experimentally realizable quasi-1D systems is extremely interesting.

In the present Letter we study a model of the quasi-1D $S=1$ antiferromagnet, which has the XXZ-type intrachain couplings and Ising-type interchain couplings. We study the model in a restricted Hilbert space $\mathcal{H}_0$ which consists only of the spin-0 defects.[12] We prove two theorems which establish the existence of a disordered ground state for the restricted model in a certain region of the parameters. Details of the proof, along with a heuristic explanation of the Haldane gap in terms of the dynamics of the spin-0 defects, will appear elsewhere.[13]

Consider a three-dimensional cubic lattice whose site is denoted as (i,α), where an integer i is the intrachain coordinate and a point α in the square lattice labels a chain. With each site we associate an $S=1$ quantum spin, and denote by $S^x_{i,\alpha}$, $S^y_{i,\alpha}$, and $S^z_{i,\alpha}$ the corresponding Pauli matrices. Our Hamiltonian is

$$H = H_c + H_s . \tag{1a}$$

The intrachain Hamiltonian H_c is

$$H_c = \sum_{i,\alpha} S^x_{i,\alpha} S^x_{i+1,\alpha} + S^y_{i,\alpha} S^y_{i+1,\alpha} + \Delta S^z_{i,\alpha} S^z_{i+1,\alpha} + D(S^z_{i,\alpha})^2 \tag{1b}$$

and the interchain coupling is

$$H_s = \epsilon \sum_{i,\langle\alpha,\beta\rangle} S^z_{i,\alpha} S^z_{i,\beta} , \tag{1c}$$

where $\langle\alpha,\beta\rangle$ denotes nearest-neighbor sites in the square lattice. The anisotropy parameters Δ and D and the interchain coupling ϵ are measured in the unit of the intrachain coupling. We require $\Delta > 0$, $\Delta \geq D$, and $\epsilon > 0$. The extensions of the results to a $(1+d)$-dimensional model and/or a model with ferromagnetic interchain couplings $(\epsilon < 0)$ are straightforward.

Our Hilbert space $\mathcal{H}_0$ is constructed by allowing any configuration of the (classical) lowest-energy excitations (spin-0 defects) from the classical ground state. To be precise, $\mathcal{H}_0$ is generated by basis states which are written as

$$0\ldots0\downarrow0\ldots0\uparrow0\ldots0\downarrow0\ldots0\uparrow0\ldots0\downarrow0\ldots0\ldots$$

in each chain. We have represented the state in the standard S^z eigenstate basis. Here $0\ldots0$ denotes a sequence of any number (including none) of the $S^z=0$ state, and the up and down spins must appear alternately. (A typical basis state looks like $\downarrow\uparrow0\downarrow\uparrow00\downarrow\uparrow\downarrow\uparrow0\downarrow$.) It is obvious that the model restricted to $\mathcal{H}_0$ is equivalent to the original model in the Ising limit $\Delta \to \infty$, but in Ref. 12 it was found that the restricted model has properties remarkably similar to the original model even when Δ is close to unity. In particular, the restricted model exhibits all the phenomena related to the Haldane gap. It is quite likely that the dynamics of the spin-0 defects provides a qualitatively correct description of the Haldane gap and the related phenomena. (This is the $S=1$ version of Haldane's picture based on the soliton dynamics.[1]) As is implicit in the following Theorem I, the restricted model fails to exhibit the XY phase which may exist in the unrestricted model when Δ is small.

Our first theorem establishes the existence of the Haldane phase and the Néel phase, and hence the existence of a transition between the two phases.

Theorem I.—In the space $\mathcal{H}_0$, the ground state of the quasi-1D Hamiltonain H has Néel order when $\Delta - D > 2$, but it is unique, has a finite excitation gap, and exponentially decaying correlation functions when $3\Delta - 2D + 8\epsilon < 2$.

The theorem also applies to the purely 1D system, where one sets $\epsilon = 0$. In this case Néel order must be understood as the order only within the chain.

Like all the rigorous theorems of this sort, the constants in the theorem are not optimal. The next theorem itself does not distinguish any concrete parameter region. Instead, it states that a quasi-1D system has a disordered ground state whenever the corresponding purely 1D system is disordered and the interchain couplings are sufficiently small.

Theorem II.—Consider a purely 1D Hamiltonian H_c with some Δ and D. Suppose that, in the space analogous to $\mathcal{H}_0$, the ground state of H_c is unique, has a finite energy gap E_1, and exponentially decaying correlation functions with the correlation length m^{-1}. Then, in the space $\mathcal{H}_0$, the ground state of the quasi-1D Hamiltonian H is also unique, has a finite excitation gap, and exponentially decaying correlation functions, when Δ and D are the same as the 1D case, and ϵ satisfies $\epsilon \leq \text{const} \times m^2 E_1^2 (m+E_1)^{-1}$.

It is crucial that the above theorem applies to a model arbitrarily close to the critical one, once we know that its 1D counterpart is in the Haldane phase. In this sense Theorem II is stronger and more important than Theorem I. Gómez-Santos[12] argued that the $S=1$ XXZ chain in the space $\mathcal{H}_0$ has a disordered ground state for $\Delta \leq \Delta_c$, where $\Delta_c \cong 1.125$ for $D=0$. If we take this conclusion as granted, the above theorem ensures that, in the same parameter region, the quasi-1D system with sufficiently small interchain couplings also has a disordered ground state.

We believe that these two theorems provide rather strong support to the conjecture that there is a quasi-1D system which does not order even at $T=0$. It is quite interesting to address the same question experimentally by investigating the low-temperature behavior of NENP. Theoretically, an interesting remaining problem would be to prove a theorem corresponding to Theorem II in the unrestricted Hilbert space.

In the rest of the Letter we sketch the main ideas used in the proof. The proof is based on an exact mapping of the quantum ground state onto an equilibrium state of a classical ferromagnetic Ising model in four dimensions. Our mapping makes use of the standard expansion of the operator e^{-TH} combined with a kind of dual transformation.

We first consider H in a finite lattice.[14] Note that the whole lattice can be decomposed into even and odd sublattices. Let the classical ground state $|C\rangle$ be the state where spins in the even sublattice have $S^z=1$ and spins in the odd sublattice have $S^z=1$. It can be proved that the ground state (in a finite volume) has a nonvanishing overlap with $|C\rangle$. We evaluate $\langle C|e^{-TH}|C\rangle$ by expanding the exponential by the Lie formula as

$$\langle C|e^{-TH}|C\rangle = \lim_{N\to\infty}\langle C|\left[\prod_{i,\alpha}\left(1-\frac{S^+_{i,\alpha}S^-_{i+1,\alpha}}{2N}-\frac{S^-_{i,\alpha}S^+_{i+1,\alpha}}{2N}\right)e^{-H^z/N}\right]^{NT}|C\rangle, \quad (2)$$

where

$$H^z=\sum_{i,\alpha}[\Delta S^z_{i,\alpha}S^z_{i+1,\alpha}+D(S^z_{i,\alpha})^2]+\epsilon\sum_{i,\langle\alpha,\beta\rangle}S^z_{i,\alpha}S^z_{i,\beta}$$

and $S^{\pm}_{i,\alpha}=(S^x_{i,\alpha}\pm S^y_{i,\alpha})/2$. NT is chosen to be an integer. We denote by $Z_{T,N}$ the quantity inside the limit of (2). We rewrite $Z_{T,N}$ as

$$Z_{T,N}=\sum_{\sigma_\tau(\tau=1,2,\dots,NT-1)}\langle C|O_N|\sigma_1\rangle\prod_{\tau=1}^{NT-2}\langle\sigma_\tau|O_N|\sigma_{\tau+1}\rangle\langle\sigma_{NT-1}|O_N|C\rangle,$$

where O_N is the operator inside the square brackets in (2), and each $|\sigma_\tau\rangle$ is summed over all the basis states of $\mathcal{H}_0$. As usual,[15] the above expression of $Z_{T,N}$ can be regarded as the partition function of a classical spin system. Here we further make use of the special characters of the space $\mathcal{H}_0$ to construct an Ising model on a dual lattice.

Since each basis state of $\mathcal{H}_0$ can be fully determined by specifying the positions of the $S^z=0$ spins (and the boundary conditions), one can regard a collection $(\sigma_1,\dots,\sigma_{NT-1})$ as describing a space-time configuration of the zero spins in $1+2+1$ dimensions. Here τ is interpreted as the (imaginary) time coordinate. Figure 1 shows the $(1+1)$-dimensional section of a typical space-time configuration, where the horizontal and the vertical axes correspond to the intrachain coordinate i and the temporal coordinate τ, respectively. Note that the zeros can propagate, and be created and annihilated in pairs, within this $(1+1)$-dimensional subspace-time.

Now we want to identify the trajectories (i.e., the lines in Fig. 1) formed by zeros with Peierls contours (i.e., boundaries separating up spins and down spins) of a ferromagnetic Ising model on a dual lattice. We consider the dual lattice $\{(k,\alpha,\tau)\}$, where α and τ are unchanged, and the intrachain coordinate is k which lies in the middle of i and $i+1$. We find that there is a one-to-one correspondence between the space-time configurations of zeros and the Peierls contours (within subspace-time with fixed α) of an Ising model on the dual lattice with positive boundary conditions. Moreover, when we take the following ferromagnetic Hamiltonian, we see that the quantum-mechanical weight associated with each configuration of zeros and the statistical-mechanical weight associated with the corresponding set of Peierls contours also coincide exactly:

$$-H_{\text{Ising},N}=\sum_{k,\alpha,\tau}\frac{\ln N}{2}\sigma_{k,\alpha,\tau}\sigma_{k,\alpha,\tau+1}+\frac{\Delta-D}{2N}\sigma_{k,\alpha,\tau}\sigma_{k+1,\alpha,\tau}+\frac{\Delta}{4N}\sigma_{k,\alpha,\tau}\sigma_{k+2,\alpha,\tau}+\sum_{k,\langle\alpha,\beta\rangle,\tau}\frac{\epsilon}{4N}(\sigma_{k,\alpha,\tau}+\sigma_{k+1,\alpha,\tau})(\sigma_{k,\beta,\tau}+\sigma_{k+1,\beta,\tau}).$$

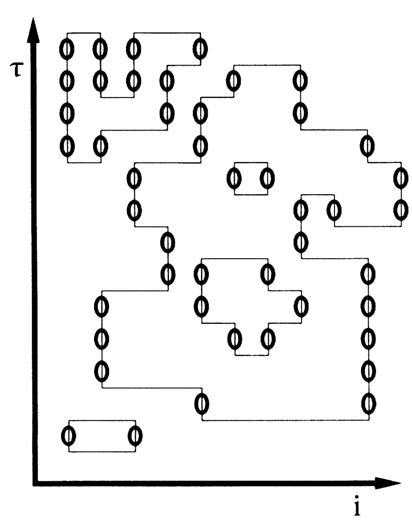

FIG. 1. The $(1+1)$-dimensional section of a typical space-time configuration of zero spins. We identify the trajectories formed by the zeros with Peierls contours in an Ising model on the dual lattice.

Thus we have that $Z_{T,N}=\text{Tr}\{\exp(-H_{\text{Ising},N})\}$.

We also find the correspondences between the spin variables $S^z_p=\pm(\sigma_{p-1/2}+\sigma_{p+1/2})/2$, where $p=(k,\alpha,\tau)$ is a site in the dual lattice and the sign takes a plus or minus depending on whether p is in the even or odd sublattice. $p\pm\frac{1}{2}$ are abbreviations of the sites $(k\pm\frac{1}{2},\alpha,\tau)$ in the original lattice. It is essential in the present approach that the quantum spin state is expressed by a local combination of the Ising spins.

Then it follows that any local operator A on $\mathcal{H}_0$ can be expressed in terms of a local combination of the Ising spins, and the ground-state expectation value of A is expressed as a statistical-mechanical expectation value of the Ising model. Then we can make use of sophisticated techniques of modern rigorous statistical mechanics to prove the desired theorems.

In a general ferromagnetic Ising model, Fisher[16] proved the following self-avoiding-walk bound

$$\langle\sigma_p\sigma_q\rangle\le\sum_{\omega:p\to q}\prod_{i=0}^{M-1}\tanh(J_{p_ip_{i+1}}),$$

where the summation is taken over all the walks $\omega=\{p_0, p_1, \ldots, p_M\}$ with $p_0=p$, $p_M=q$ which pass through each bond at most once. We use this bound to prove the second half of Theorem I. In our case $p,q,\ldots$ are sites in the four-dimensional space-time (dual) lattice, and the interaction J_{pq} is determined from the Ising Hamiltonian $H_{\text{Ising},N}$. When the interactions J_{pq} are sufficiently small, the right-hand side of the Fisher bound becomes a convergent sum, and we get exponentially decaying upper bounds for the correlation functions. To prove a meaningful result in the quantum system, however, these bounds and the exponential decay rates must survive the limit $N\to\infty$. This is not easy because the interaction in the temporal direction grows as $\ln N$. Again following Fisher, we make full use of the self-avoiding nature of ω and the fact that the couplings in space directions decrease as $1/N$ to get bounds uniform in N. They lead to the following bounds for the quantum-mechanical correlation functions which hold for arbitrary local operators A and B:

$$|\omega(AB)-\omega(A)\omega(B)|\le C_1\exp[-\text{dist}(A,B)/\xi_0]\,, \quad (3a)$$

$$|\omega(e^{TH}Ae^{-TH}B)-\omega(A)\omega(B)|\le C_2\exp(-tE_0)\,. \quad (3b)$$

Here

$$\omega(A)=\lim_{L\to\infty}\lim_{T\to\infty}\frac{\langle C|e^{-TH/2}Ae^{-TH/2}|C\rangle}{\langle C|e^{-TH}|C\rangle}$$

is the ground-state correlation function in the infinite-volume limit. $\text{dist}(A,B)$ is the distance between the domains of the operators A and B. The constants C_1,C_2 depend only on the operators A,B. ξ_0 gives the upper bound for the correlation length, and E_0 the lower bound for the excitation gap.

The first part of Theorem I is easier. First, by the Griffiths inequality, we bound from below the desired order parameter by that of an Ising model on the square lattice whose horizontal and vertical nearest-neighbor interactions are $J_H=(\Delta-D)/N$ and $J_V=\ln N/2$, respectively. Then we get the desired result from the exact solution of the 2D Ising model.

In Theorem II, it is assumed that the above bounds (3a) and (3b) hold for the model with $\epsilon=0$. We want to prove that after adding small ϵ, the bounds (3a) and (3b) remain valid with slightly modified decay rates. This is quite a delicate problem because we have to rigorously rule out the possibility of a first-order transition and, even more difficult, again have to get the estimates which survive the $N\to\infty$ (and the infinite-volume) limits. The corresponding problem (without the $N\to\infty$ difficulty) in the classical spin systems was first solved by Simon[17] who made use of a correlation inequality known as the Simon-Lieb inequality.[18] Even in the present case, by extending Simon's method carefully, we get the bounds uniform in N.

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